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Review

Enriched Crystal Scintillators for 2β Experiments

Oksana G. Polischuk

Institute for Nuclear Research of NASU, 03028 Kyiv, Ukraine; polischuk@kinr.kiev.ua

Abstract: The investigation of 2β decay is an important issue in modern physics, allowing the test of the Standard Model of elementary particles and the study of the nature and properties of neutrinos. The crystal scintillators, especially made of isotopically-enriched materials, are powerful detectors for 2β decay experiments thanks to the high radiopurity level and the possibility to realize the calorimetric "source = detector" approach with a high detection efficiency. For the moment, the 2ν2β processes have been observed at the level of 10^{19}–10^{24} years with enriched crystals; the sensitivity to the 0ν mode have reached the level of 10^{24}–10^{26} years in some decay channels for different nuclides allowing one to calculate the upper limits on the effective mass of the Majorana neutrino at the level of 0.1–0.6 eV. The paper is intended to be a review on the latest results to investigate 2β processes with crystal scintillators enriched in ^{48}Ca, ^{106}Cd, and ^{116}Cd.

Keywords: enriched crystals; scintillators; double beta decay

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1. Introduction

2β decay is the rarest process observed in nature in which nuclei with atomic mass A spontaneously change their charge Z by two units: (A,Z) → (A,Z ± 2) involving the simultaneous emission of two electrons or positrons. The 2ν mode (the transition accompanied by two (anti)neutrinos (2ν2β)) is allowed by the Standard Model (SM), and can be expressed for nucleus as ${}^{A}_{Z}X \rightarrow {}^{A}_{Z+2}X + 2e^- + 2\overline{\nu}$ (${}^{A}_{Z}X \rightarrow {}^{A}_{Z-2}X + 2e^+ + 2\nu$). The probability of the decay is inversely proportional to the half-life $\left[T^{2\nu}_{1/2}\right]^{-1} = G_{2\nu}\left|M^{2\nu}\right|^2$, where $G_{2\nu}$ is a phase space factor and $M^{2\nu}$ is the nuclear matrix element (NME) [1]. While the $G_{2\nu}$ can be precisely calculated, the calculations of NMEs are complex and depend on the selected nuclear model [2–4]. The half-life (or the lower limit on the half-life) relative to different channels of 2β decay can be estimated using the following formula:

$$T_{1/2}(\text{or } \lim T_{1/2}) = N \times \ln 2 \times \eta_{\text{det}} \times \eta_{\text{sel}} \times t/S(\text{or } \lim S), \quad (1)$$

where N is the number of nuclei in the sample, η_{det} is the detection efficiency of the decay (the ratio of the number of events in the simulated distribution to the number of generated events), η_{sel} is the efficiency of events selection, t is the time of measurement, and S is the number of events of the effect searched for, which can be excluded at a given confidence level. The precise measurements of $T_{1/2}$ for 2ν2β decay for different nuclei could help to specify the methods and parameters of theoretical calculations of NMEs.

After more than 75 years of investigations, the 2ν2β decay to the ground state has been observed for 11 nuclides (^{48}Ca, ^{76}Ge, ^{82}Se, ^{96}Zr, ^{100}Mo, ^{116}Cd, ^{128}Te, ^{130}Te, ^{150}Nd, ^{136}Xe, and ^{238}U) with half-lives in the range of 10^{18}–10^{24} years [5] in direct, geochemical, and radiochemical experiments. The average values of the half-lives relative to 2ν2β decay are shown in Figure 1 (black squares). The 2ν2β decay to the first 0^+ excited level of the daughter nucleus in ^{100}Mo and ^{150}Nd has also been detected with half-lives of $T_{1/2} \sim 10^{20}$ years [5,6]. The processes of 2β decay with nuclear charge decreasing have been observed only for ^{124}Xe [7] and ^{78}Kr [8] in direct experiments, but also for ^{130}Ba in geochemical experiments [9,10].

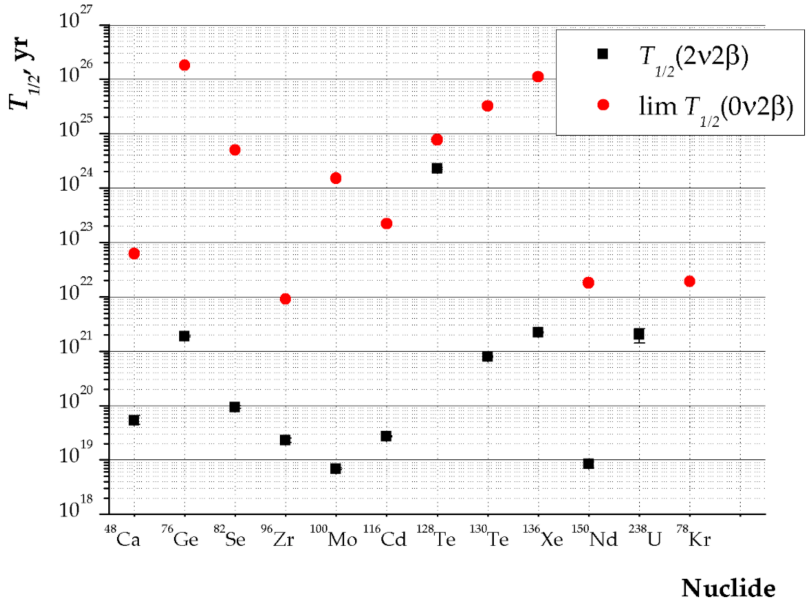

Figure 1. The average values of the half-lives (in years) relative to 2ν2β decay (black squares) [5] and the highest experimental values on the lower limits of the half-lives related to 0ν2β decay for different isotopes (red circles, see text). Errors are given at 68% C.L., while the lower limits are given at 90% C.L. 2ν2β decay for the ^{128}Te isotope is observed only in the geochemical experiment.

The neutrinoless 2β decay (0ν2β), a transformation which changes the charge of a nucleus by two units without (anti)neutrino emission ($^{A}_{Z}X \rightarrow {}^{A}_{Z+2}X + 2e^{-}$), is forbidden in the SM due to the lepton number violation. The process has not been observed yet, only the most stringent half-life limits have been set at the level of 10^{25}–10^{26} years (red circles in Figure 1). Thanks to the observation of neutrino oscillations, the neutrino is supposed to be a particle with non-zero mass [11,12]. While the oscillation experiments are sensitive to the squared neutrino mass eigenstates difference, 0ν2β decay is a possible way to test the lepton number conservation in weak interactions, clarify the nature of neutrino (Dirac or Majorana particle), and its observation would provide the information about the neutrino mass scale [13–24]. The investigation of neutrinoless electron capture with positron emission (0νεβ^{+}) and neutrinoless double positron emission (0ν2β^{+}) is one of possible ways to refine the mechanism of the decay due to the light ν mass or due to the right-handed currents admixture [25]. The development of experimental methods for the research of different 2β isotopes is highly required because of big uncertainties of the theoretical estimations for 0ν2β decay probability [26].

The highest experimental values on the lower limits of the half-lives related to 0ν2β decay for ^{48}Ca [27], ^{76}Ge [28], ^{78}Kr [8], ^{82}Se [29], ^{96}Zr [30], ^{100}Mo [31], ^{116}Cd [32], ^{128}Te [33,34], ^{130}Te [35], ^{136}Xe [36], and ^{150}Nd [37] are shown in Figure 1 (red circles). The sensitivity of the experiments with enriched crystals reached the level of 10^{24}–10^{26} years for some 2β channels for different nuclides that are close to the theoretical estimations allowing to calculate the upper limits on the effective mass, ‹m_{υ}›, of the Majorana neutrino at the level of ‹m_{υ}› ~ 0.1–0.6 eV; see [38] for a review and the results obtained in the experimental works [28,35,36].

Crystal scintillators are widely utilized in particle physics, in particular, for search for 2β decay, dark matter (DM) particles, and the study of rare nuclear decays (alpha, beta, cluster decays, etc.); see [39] and references therein. The sensitivity of the 2β decay experiment depends on the following values [40,41]:

$$\lim T_{1/2} \sim \varepsilon \cdot \delta \sqrt{\frac{m \cdot t}{FWHM \cdot Bg}}, \quad (2)$$

where ε is the detection efficiency, δ is the abundance of the investigated isotope in the sample, m and $FWHM$ (full width at half maximum) are the total mass and the energy resolution of the detector, t is the time of measurement, and Bg is the background rate in the region of interest for the searched effect. The $\lim T_{1/2}$ value directly is proportional to the detection efficiency and the enrichment of the isotope, while the other parameters are under the square root. So the development of isotopically enriched materials that contain the maximum value of the investigated isotopes for 2β decay experiments is strongly required. Based on Equation (2), the crystal scintillators are considered as powerful detectors to search for 2β processes due to the following general advantages:

- realization of the "source = detector" approach by detecting events whose origins are within the crystal; in this case the detection efficiency for searched effects could reach 80−90%;
- possibility of isotope enrichment for many of the crystals [42]; some enrichment techniques (like the gas centrifuge method) are not so expensive, while others (for example, the multi-channel counter current electrophoresis (MCCCE) or laser separation) are much complicated and expensive;
- an ability to increase the sensitivity of the experiments by the use of big-size crystals [43,44] or large array of crystals;
- low level of the internal radioactive contamination (typically the crystals for low background measurements have contamination by ^{226}Ra and ^{228}Th on the level of ~(0.001−0.1) mBq/kg;
- possibility of the development of radiopure scintillators;
- the good pulse-shape discrimination factor;
- very good operating stability (for years).

Furthermore, since the investigation of such rare processes requires special conditions such as a low level of external radioactive background, the experiments are carried out mostly at the low-background underground laboratories.

In order to investigate the 0ν2β decay, isotopes with the high decay energy are favored due to the increasing probability of the decay (as E^5), but also the sharp decreasing of the natural radioactivity after 2615 keV (energy of γ quanta of ^{208}Tl from ^{232}Th chain) and lower cosmogenic activations at a high-energy region; this advantage simplifies the problem of induced background. Another challenge in the research is a possible contribution from the 2ν2β mode to the 0ν2β decay region. A good energy resolution of the crystal scintillators is preferable to overcome this problem. It could be provided by using enriched crystals as cryogenic detectors for which effective particle discrimination by the simultaneous detection of phonon and scintillation signals is inherent. The most perspective low-temperature scintillating detectors for the 2β decay searches are the scintillators enriched by ^{100}Mo: Calcium molybdates ($Ca^{100}MoO_4$) [45,46], lithium molybdates ($Li^{100}MoO_4$) [47,48], and zinc molybdates ($Zn^{100}MoO_4$) [49,50]. ^{100}Mo is one the most promising 2β isotope thanks to a high natural isotopic abundance δ = 9.8% [51] (it can also be enriched using relatively inexpensive ultra-speed centrifuge technology) and its large energy release $Q_{2\beta}$ = 3034.40(17) keV [52] above the edge of natural radioactivity. In particular, cryogenic scintillation bolometers with molybdate crystals enriched in ^{100}Mo with a relatively high energy resolution (a few keV at $Q_{2\beta}$) are accepted for large-scale projects of the new generation to search for 0ν2β decay aiming at a half-life sensitivity $\geq 10^{27}$ years (with sensitivity to neutrino mass at the level of the inverse scheme of neutrino mass states 0.02–0.05 eV), like AMORE (Advanced Mo-based Rare process Experiment) [53] and CUPID (CUORE Upgrade with Particle Identification) [54]. The best up-to-date sensitivity to 0ν2β decay in ^{100}Mo was reached: $T_{1/2}^{0\nu 2\beta} > 1.5 \times 10^{24}$ years at 90% confidence level (C.L.) which corresponds to the effective Majorana mass of neutrino $\langle m_{2\beta} \rangle$ < (0.31–0.54) eV [31]). Some other enriched crystals using bolometers ($^{130}TeO_2$ [55], $Zn^{92}Se$ [29], $^{116}CdWO_4$ [56]) are also a perspective for high-sensitivity 0νββ decay searches.

The paper focuses on some isotopes for 2β processes investigation that could be used as crystal scintillators at room temperature and fall under most demands listed above (^{48}Ca, ^{106}Cd, and ^{116}Cd). The review on the latest results for these isotopes is given. Since the mass of these detectors does not exceed a few kilograms, they could be used mostly for the 2ν2β decay search. The precise measurements of the 2ν mode is important for NME calculations because there are still large differences in the literature between the NME values.

2. Crystal Growth and Radiopurity of Crystal Scintillators

2β experiments, at different stages over the world, running or being under development, require crystal scintillators with as low as possible radioactive contamination ~(0.1–1) μBq/kg by U/Th daughters (the secular equilibrium of the U/Th chains could be broken and should be considered separately), ^{40}K, surface contamination and cosmogenic activation by ^{110m}Ag, ^{56}Co, ^{88}Y, and ^{88}Zr, radioactive isotopes of rare-earth elements could also be found in the crystals [57–59], as well as anthropogenic ^{90}Sr-^{90}Y and ^{137}Cs caused by the Chernobyl (1986), and the Fukushima Daiichi (2011) nuclear disasters [60]. As additional contamination in low-background experiments, one should consider also an initial activity of the different compounds of the crystals (for example, 2β activity of ^{48}Ca, ^{100}Mo, and ^{116}Cd can contribute in the regions of the interest for the rare processes of these isotopes). The main objectives of different projects are investigation and R&D (research and development) of deeply radiopure crystal scintillators for low-background measurements to investigate 2β decay and search for DM. R&D of very low radioactive scintillators calls for a deep collaboration of chemists and crystal growers with low radioactivity experts. Crystal producers should pay more attention on radioactive contamination and behavior of tiny bulk and surface pollutions because even a small fraction of some radioactive impurities can play a crucial role in the low-counting measurements. It is important to realize a fast screening of crystal samples to investigate the radioactivity of all the crystal growth equipment, equilibrium issues, surface contamination, cosmogenic activation, and radon deposit. A very low radioactive contamination, required by the next generation of 2β decay experiments, can be measured by using a low background "source = detector" approach. The method can provide the sensitivity at the level of μBq/kg for ^{40}K, ^{137}Cs, ^{228}Th, and U/Th daughters [61]. The efforts in the research of new materials and purification techniques as well as the improvement of the crystal growth procedures allows a wide choice among inorganic scintillators for a variety of uses and reaching progress in 2β investigations.

The radiopurity level of isotopically enriched crystals is typically lower than it is required for the low-background experiments, whose requirements on radiopurity level are ~(0.1–1) μBq/kg (for example, the total internal α activity in the cadmium tungstate crystals enriched by $^{106/116}$Cd is ~(2–3) mBq/kg, while for natural crystal it is ~(0.2–0.3) mBq/kg). The choice of the best purification and production cycle of enriched scintillators includes:

(1) minimization of the initial materials and possible radioactive components of the growing set-ups;
(2) a test of the radiopurity level of the detectors;
(3) a study of the effect of double crystallization, development of protocol for surface treatment;
(4) an application of methods to purify enriched isotopes by using combination of chemical (recrystallization) and physical (vacuum distillation, filtration, zone melting) approaches;
(5) minimization of cosmogenic/neutrons activation (the production line and transportation of the crystals to the experiment site should be provided with the safest way, by ground-based) and radioactive background deposit from radon.

For the moment, one of the best techniques of enriched crystal production is the low-thermal gradient Czochralski crystal growth method (LTG CZ) [62] which provides large volume radiopure crystal scintillators with an excellent optical quality, relatively

high yield of crystalline boules (for example, ~87% of the initial charge for $CdWO_4$, and an acceptable level of irrecoverable losses of expensive initial enriched materials (~2% for $CdWO_4$).

The improvement of a radiopurity level of enriched crystal scintillators could also be achieved by recrystallization [63,64]. For example, comparing the data of experiments for $^{116}CdWO_4$ crystals, before and after recrystallization, one can conclude that segregation coefficient for K, Th, and especially of Ra is very low during crystal growth. It gives a strong signature that the radioactive contamination of the crystals by ^{40}K, ^{228}Th, and ^{226}Ra can be substantially reduced by recrystallization. The total α activity due to the U/Th chains after recrystallization has been improved by a factor ~3; the contamination by thorium (^{228}Th) has been reduced by one order of magnitude to the level of 0.01 mBq/kg (see Table 1). The same behavior was also discovered for molybdates (including crystals enriched in ^{100}Mo) [50] and zinc tungsten crystals [64]. The development of other crystals by LTG CZ (for example, calcium and lead molybdates ($CdWO_4$, $PbMoO_4$) is in progress.

Table 1. Radioactive contamination of the ^{116} $CdWO_4$ crystal before and after recrystallization [64].

Chain	Nuclide	Activity, mBq/kg	
		Before Recrystallization	After Recrystallization
^{232}Th	^{232}Th	0.13 (7)	0.03 (2)
	^{228}Th	0.10 (1)	0.010 (3)
^{238}U	^{238}U	1.8 (2)	0.8 (2)
	^{226}Ra	$\leq$0.1	$\leq$0.015
	$^{234}U + ^{230}Th$	0.6 (2)	0.4 (1)
	^{210}Po	1.6 (2)	0.4 (1)
Total α		4.44 (4)	1.62 (4)

3. Calcium Fluoride Crystal Scintillators Enriched in ^{48}Ca

The ^{48}Ca is the lightest nucleus among potential 2β decay isotopes with the highest energy release ($Q_{2\beta}$ = 4268.08(8) keV [52]; a simplified ^{48}Ca decay scheme is shown in Figure 2), promising theoretical calculations, and, despite the low natural isotopic abundance (δ = 0.187(21)%), a possibility for enrichment up to almost 100%.

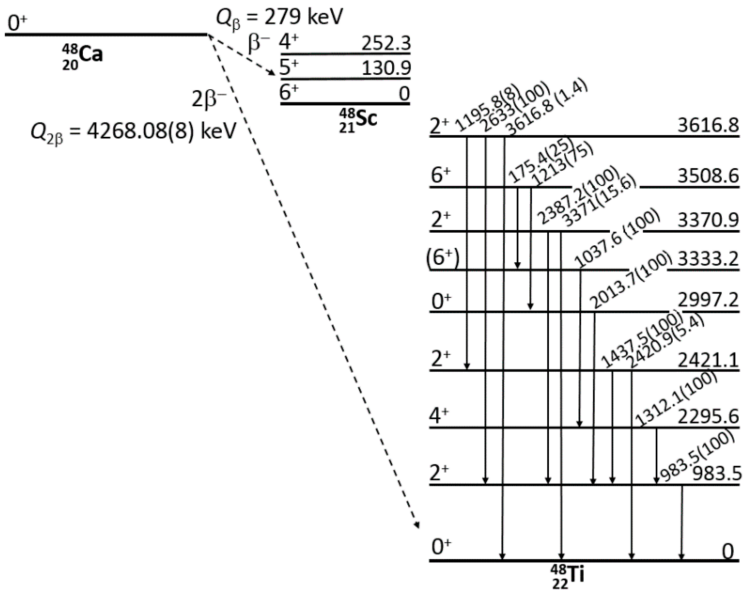

Figure 2. A simplified ^{48}Ca decay scheme. Energies of the excited levels and of the emitted γ quanta are in keV. The relative intensities of γ quanta are given in parentheses [52].

The first experiment with europium-activated calcium fluoride crystal scintillator, $CaF_2(Eu)$, enriched in ^{48}Ca up to 96.59% has been performed in 1966 at the Brookhaven National Laboratory with the aim to search 2β processes in ^{48}Ca. The detector of 2 cm × 1.8 cm with a total mass of ^{48}Ca about 10.6 g worked in anticoincidence mode with a plastic scintillator shield. Only the limit on 0ν2β decay was set as $T_{1/2}^{0\nu2\beta} > 2 \times 10^{20}$ years [65].

There were few attempts with CaF_2 crystals with a natural abundance of ^{48}Ca to investigate 2β processes in ^{48}Ca. The limits on 0ν2β decay were established as $T_{1/2}^{0\nu2\beta} > 1.4 \times 10^{22}$ years and $T_{1/2}^{0\nu2\beta} > 5.8 \times 10^{22}$ years at 90% C.L. with the help of CaF_2 and $CaF_2(Eu)$ scintillators in [66,67], respectively.

In addition, the CANDLES experiment (CAlcium fluoride for the study of Neutrinos and Dark matters by Low Energy Spectrometer) is in progress at the Kamioka underground observatory (2700 m of water equivalent), Japan. The use of enriched $^{48}CaF_2(Eu)$ crystals up to 2% (and then to 50%) by ^{48}Ca is expected; the collaboration has been developing the ^{48}Ca enrichment with MCCCE or laser separation and the possibility of large-scale enrichment is still under investigation. Meanwhile, for the first stage, 96 CaF_2 cylindrical crystals (of the diameter and length, Ø5 cm × 5 cm, with natural abundance by ^{48}Ca) with a total mass of 305 kg are using as scintillating bolometers. A liquid scintillator provides an active 4π shield. The energy resolutions for the detectors at the Q-value of ^{48}Ca is ~2.6% (a better energy resolution is required to reduce an irremovable background 2νββ background), and of 4.0–4.5% at 1460.8 keV and 2.9–3.3% at 2614.5 keV. The new limit on 0ν2β decay was set as $T_{1/2}^{0\nu2\beta} > 6.2 \times 10^{22}$ year at 90% C.L. [27,68].

4. Cadmium Tungstate Crystal Scintillators Enriched in ^{106}Cd

^{106}Cd is one of the most appealing candidates for $2\beta^+$ decay search with natural abundance δ = 1.245(22)% with a possibility of enrichment up to 100% by using relatively inexpensive gas centrifugation techniques. Energy release is high enough, $Q_{2\beta}$ = 2775.39(10) keV [52], for different processes like double positron emission decay ($2\beta^+$), electron capture with positron emission ($\varepsilon\beta^+$), and double electron capture (2ε). A simplified ^{106}Cd decay scheme is shown in Figure 3. The isotope is also interesting by its possible near resonant neutrinoless 2ε captures ($0\nu2\varepsilon$) from different shells (K, L_1, L_3) to the excited level of ^{106}Pd (2718 keV for 0ν2K, 2741 keV for $0\nu KL_1$ and 2748 keV for $0\nu KL_3$). For some modes of the 2β processes, theoretical $T_{1/2}$ are promising 10^{20}–10^{22} years (ground state (g.s.) to g.s.) and reachable by modern low-counting techniques [69].

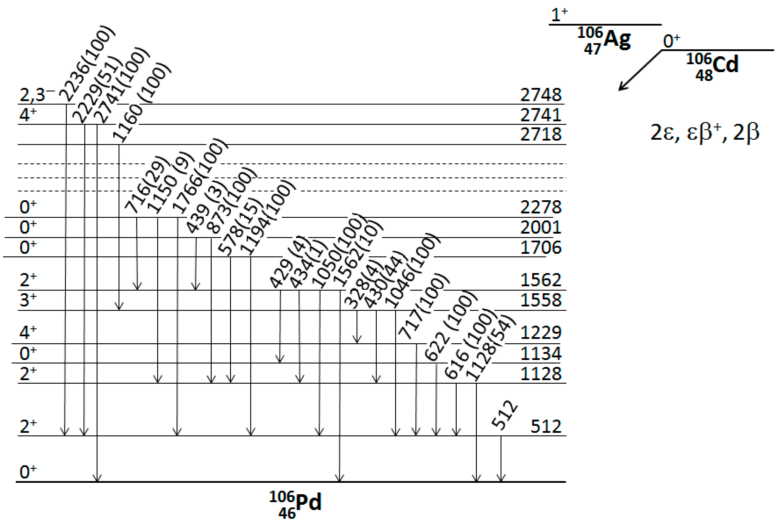

Figure 3. A simplified ^{106}Cd decay scheme [70]. Energies of the excited levels and of the emitted γ quanta are in keV. The relative intensities of γ quanta are given in parentheses. For the levels of ^{106}Pd with energies 2718 keV, 2741 keV, and 2748 keV the resonant 0ν2EC decays are possible.

$^{106}CdWO_4$ crystal enriched in ^{106}Cd up to 66.4% have been developed for the high-sensitivity experiments to search for 2β decay of ^{106}Cd [71]. Cadmium samples (including enriched ones) were deeply purified by a vacuum distillation technique with filtering on getter filters and cadmium tungstate compounds were synthesized from solutions [72,73]. The isotopic composition was precisely measured by thermal ionization mass-spectrometry (TIMS). The crystal boule of $^{106}CdWO_4$ with a mass of 231 g was grown by the low-thermal-gradient Czochralski technique [62,74]. Then it was cut and the surface has been polished to improve the light collection (see Figure 4). The final total mass became 216 g.

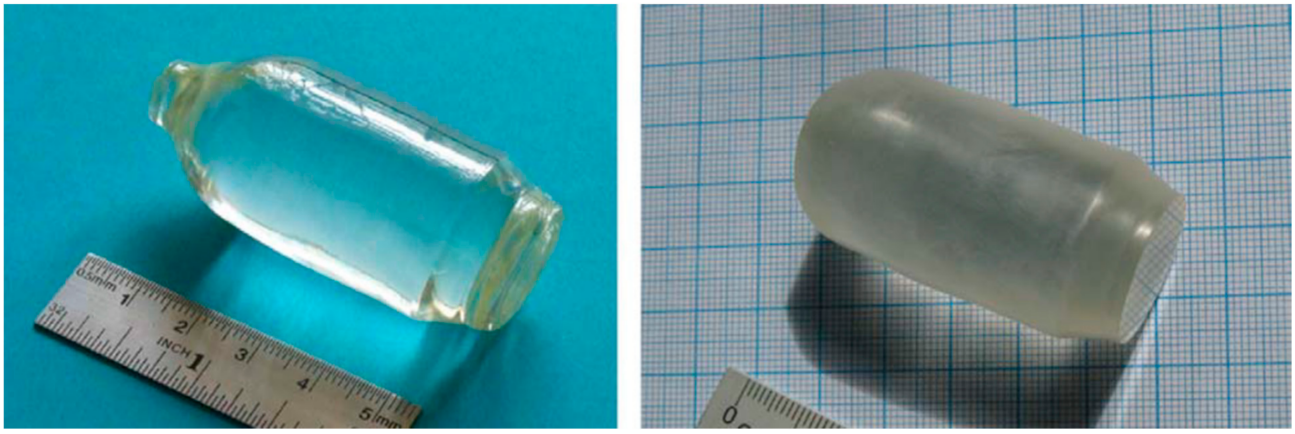

Figure 4. The crystal boule of ^{106} $CdWO_4$ (231 g) was grown by the low-thermal-gradient Czochralski technique (**left**). $^{106}CdWO_4$ crystal scintillator (216 g) with diffused surface for better light collection (**right**). The figure was taken from [71].

Few stages of experiments with the $^{106}CdWO_4$ crystal enriched in ^{106}Cd up to 66% were carried out at the Gran Sasso underground laboratory of the INFN (LNGS, Assergi, Italy) at a depth of 3600 m of water equivalent. The scintillator shows excellent optical and luminescence properties, good α/γ discrimination capability, and low levels of intrinsic contamination by the primordial ^{40}K and radionuclides from U/Th chains, anthropogenic ^{90}Sr and ^{137}Cs, cosmogenic ^{110m}Ag (in particular, <1.4 mBq/kg of ^{40}K, 0.005 mBq/kg of ^{228}Th, 0.012 mBq/kg of ^{226}Ra, and with the total α activity ~2.1(2) mBq/kg). The low energy resolution of the enriched detector (*FWHM* ~ 10% at 662 keV), which was measured with ^{22}Na, ^{60}Co, ^{133}Ba, ^{137}Cs, and ^{228}Th γ sources, could be explained by its elongated shape, which leads to a low and non-uniform collection of scintillation light [69,71,75].

At the first stage, the $^{106}CdWO_4$ scintillator (216 g) enriched in ^{106}Cd up to 66% was installed in the low-background DAMA R&D set-up. After 6590 h of data taking, new half-life limits on the 2β decay processes were set at the level of 10^{19}–10^{21} years [69].

For the second stage, a lead tungstate ($PbWO_4$) light-guide was developed from archeological lead to suppress the background owing to the radioactive contamination from the photomultiplier tube (PMT) [76,77]. The same crystal, as in the first stage, was installed in an ultra-low-background high purity germanium (HPGe) γ spectrometer GeMulti of the STELLA (SubTErranean Low Level Assay) facility [78] in a well in the center of one cryostat with four HPGe detectors (~225 cm^3 each). The scintillator was viewed through the $PbWO_4$ crystal light-guide (Ø40 mm × 83 mm) by 3-inch low-radioactive PMT Hamamatsu R6233MOD (see Figure 5). The detection of γ quanta, inherent for the most ^{106}Cd decay channels, was expected, including the annihilation γ quanta emitted in decay modes with positron emissions. Despite the protection of the crystal from the radioactive contamination of PMT, the use of archeological $PbWO_4$ crystal led to a decreasing of the energy resolution due to its non-perfect transparency [79]. The improved limits have been set after 13,085 h of data taking with respect to different 2β decay channels at the level of $T_{1/2}$ ~ (10^{20}–10^{21}) years [75]. In particular, the half-life limit on the two-neutrino electron capture with positron emission has reached the region of theoretical predictions: $T_{1/2}^{2\nu\varepsilon\beta+} > 1.1 \times 10^{21}$ years at 90% C.L (see Table 2).

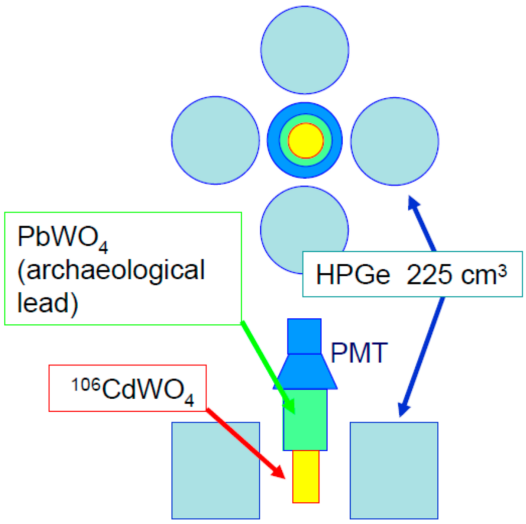

Figure 5. A scheme of the experimental GeMulti set-up with $^{106}CdWO_4$.

At the third stage, the $^{106}CdWO_4$ detector was processing in the low-background DAMA/Crys set-up in coincidences/anti-coincidences with two large volumes of $^{106}CdWO_4$ crystal scintillators in close geometry with the aim to increase the detection efficiency of γ quanta. The $^{106}CdWO_4$ crystal scintillator was viewed through the $PbWO_4$ crystal light-guide (Ø40 mm $\times$ 83 mm) by 3-inch low-radioactive PMT (Hamamatsu R6233MOD). Two cylindrical detectors from natural cadmium $CdWO_4$ (Ø70 mm $\times$ 38 mm) were viewed by two 3-inch low-background PMTs (EPI EMI9265B53/FL) through optical light-guides composed of two parts: Ultrapure quartz (Ø66 mm $\times$ 100 mm) and polystyrene (Ø66 mm $\times$ 100 mm) (see Figures 6 and 7). The passive shield consists of copper (11 cm), lead (10 cm), cadmium (2 mm), and polyethylene (10 cm) with additional flushing by high-purity nitrogen gas to suppress the radon-caused background. The amplitude, arrival time, and pulse shape of each event were recorded.

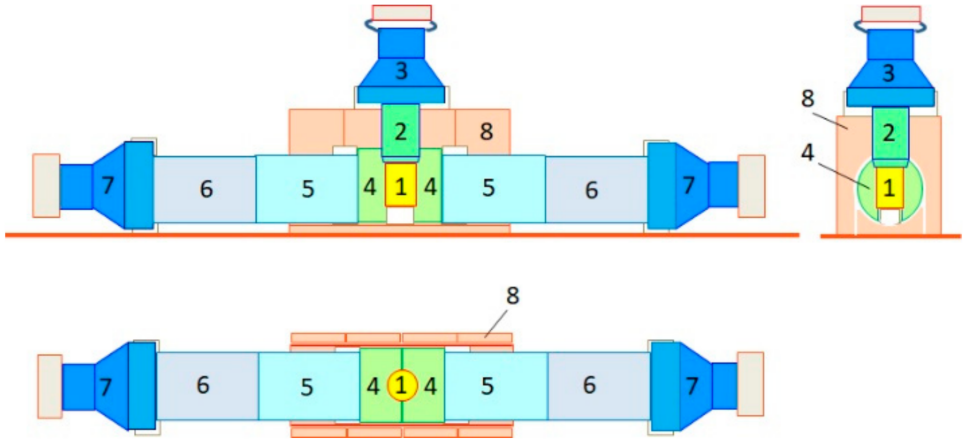

Figure 6. A scheme of the experimental set-up with $^{106}CdWO_4$. $^{106}CdWO_4$ crystal scintillator (1) is viewed through the $PbWO_4$ light-guide (2) with the help of Photomultiplier Tube (PMT) (3). Two $CdWO_4$ crystal scintillators (4) are viewed by two PMT (7) through optical light-guides composed of two parts: Ultrapure quartz (5) and polystyrene (6). The passive shield consists of copper (8—some parts are shown), lead, polyethylene, and cadmium. The figure was taken from [80].

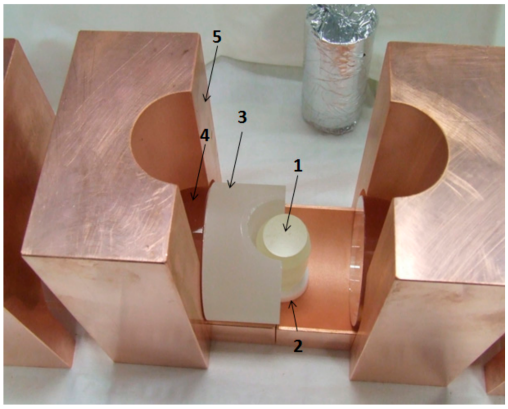

Figure 7. $^{106}CdWO_4$ crystal scintillator (1), teflon stand for the $^{106}CdWO_4$ crystal (2), $CdWO_4$ crystal scintillators (3), quartz light-guide (4), and "internal" copper shield (5).

After 26033 h of data taking there are no features that can be attributed to the processes of 2β decay of the ^{106}Cd in the experimental data. The lower limits on the half-life of ^{106}Cd relative to different channels of 2β decay were estimated by using Equation (1). The responses of the detector system to different channels of 2β decay of ^{106}Cd, as well as the contribution of radioactive contamination of the $^{106}CdWO_4$ crystal scintillator, including cosmogenic ^{110m}Ag and 2ν2β$^-$ decay of ^{116}Cd with the half-life $T_{1/2}^{2\nu2\beta} = 2.63 \times 10^{19}$ years [32], external γ quanta from the setup materials (^{232}Th, ^{238}U, and ^{40}K in the cryostat of the HPGe detector, PMT, light guide, ^{26}Al in the aluminum well of the cryostat), distribution of α particles (which passed the pulse-shape discrimination cut to select β and γ events) were simulated by using the EGSnrc package [81] with the initial kinematics given by the event generator DECAY0 [82].

The best half-life limits (as well as the theoretical estimations) on some 2β processes in ^{106}Cd are presented in Table 2 [80]. In particular, new limits on 2ε, εβ$^+$, and 2β$^+$ processes were set on the level of $T_{1/2} > (10^{20}–10^{22})$ years. The half-life limit on the 2νεβ$^+$ decay to the ground state, $T_{1/2}^{2\nu\varepsilon\beta+} > 2.1 \times 10^{21}$ years, reached the region of theoretical predictions; for 0ν2ε resonant captures the limits were set as $T_{1/2}^{0\nu2\varepsilon} > (0.35 - 2.9) \times 10^{21}$ years.

Table 2. Half-life limits on some 2β processes in ^{106}Cd. The theoretical estimations are also given.

Decay Channel, Level of ^{106}Pd (keV)	lim$T_{1/2}$(years) at 90% C.L.	
	Best Limits	**Theory**
2ν2ε, 0_1^+ 1134	$\geq 9.9 \times 10^{20}$ [80]	1.1×10^{24} [83]
0ν2ε, g.s.	$\geq 1.1 \times 10^{21}$ [69]	
2νεβ$^+$, g.s	$\geq 2.1 \times 10^{21}$ [80]	8.3×10^{20} [84] 2.7×10^{22} [83] 7.7×10^{22} [85]
2νεβ$^+$, 0_1^+ 1134	$\geq 1.1 \times 10^{21}$ [69]	1.1×10^{27} [83]
0νεβ$^+$, g.s.	$\geq 1.4 \times 10^{22}$ [80]	3.4×10^{26} [25]
2ν2β$^+$, g.s.	$\geq 2.3 \times 10^{21}$ [69]	2.4×10^{27} [83] 3.1×10^{27} [85]
0ν2β$^+$, g.s.	$\geq 5.9 \times 10^{21}$ [80]	4.8×10^{27} [25] $(1.9–3.2) \times 10^{27}$ [86]
Res. 0ν2ε, 2718	$\geq 2.9 \times 10^{21}$ [80]	
Res. 0ν2ε, 2741	$\geq 9.5 \times 10^{20}$ [69]	
Res. 0ν2ε, 2748	$\geq 1.4 \times 10^{21}$ [69]	

After almost 1 year of data taking there are no features that can be attributed to the processes of 2β decay of the ^{106}Cd in the experimental data. The sensitivity of the experiment for the different channels of 2β decay of ^{106}Cd is expected to be 10^{21}–10^{22} years during five years of measurements that is near the theoretical predictions (see Table 2). The measurements and data analysis are in progress.

5. Cadmium Tungstate Crystal Scintillators Enriched in ^{116}Cd

^{116}Cd is a promising isotope to search for 0ν2β decay thanks to its high energy release ($Q_{2\beta}$ = 2813.49(13) keV) [52], isotopic abundance (δ = 7.49(18)%), possible enrichment in large amount (despite the relatively high price), and promising theoretical calculations for 2β processes. A simplified decay scheme of ^{116}Cd is shown in Figure 8.

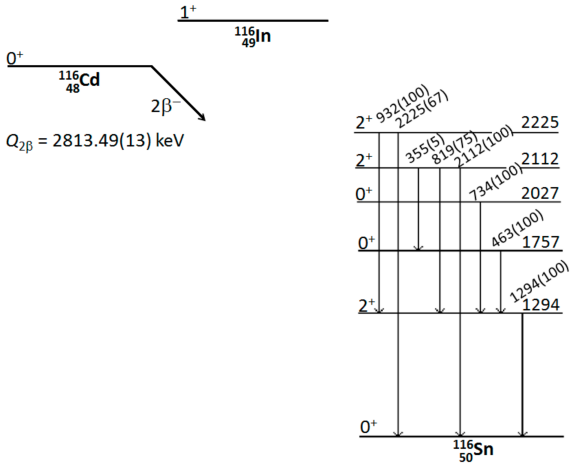

Figure 8. Simplified decay scheme of ^{116}Cd [87]. Energies of the excited levels and of the emitted γ quanta are in keV. The relative intensities of γ quanta are given in parentheses.

$CdWO_4$ crystal scintillators have excellent scintillation properties, low level of internal radioactive contamination, good particle discrimination ability, and could be used in the "source = detector" approach. In particular, the light output of the detectors is ~20% in comparison with NaI(Tl), the wavelength of emission maximum is 480 nm with principal decay time 13 μs.

For the first measurement, the $^{116}CdWO_4$ crystal scintillators, enriched in ^{116}Cd to 83%, have been developed for the 2β decay investigation [88]. Four $^{116}CdWO_4$ crystals with a total mass of 330 g were installed inside a set-up at the Solotvina Underground Laboratory (Ukraine). They were viewed by a low-background 5-inch PMT through a light-guide (Ø10 cm × 55 cm) which was composed of 25 cm of high purity quartz and 30 cm of plastic scintillator. The active shield consisted of 15 $CdWO_4$ crystal scintillators with a total mass of 20.6 kg. The set-up is described in details in [89]. The pulse-shape discrimination technique was applied to obtain the energy spectra of β(γ) events with the aim to investigate different modes of 2β decay in ^{116}Cd. The results of the experiment can be found in [90]. In particular, the half-life value on the 2ν2β decay of ^{116}Cd was set as $T_{1/2}^{2\nu2\beta} = 2.9^{+0.4}_{-0.3} \times 10^{19}$ years, and the new half-life limit on the 0ν2β decay of ^{116}Cd has been established as $T_{1/2}^{0\nu2\beta} > 1.7 \times 10^{23}$ years at 68% C.L. Other 2β processes in ^{106}Cd, ^{108}Cd, ^{114}Cd, ^{180}W, and ^{186}W nuclei were restricted at level 10^{17}–10^{21} years [89,90].

For the second experiment, called Aurora, to investigate 2β processes in ^{116}Cd, new $^{116}CdWO_4$ crystals enriched in ^{116}Cd up to 82% have been developed [91]. A large-mass boule (~1.9 kg) of the enriched $^{116}CdWO_4$ crystal (see Figure 9) was cut on three cylindrical parts; two of them (580 g and 582 g) were used for the experiment. The scintillating detectors shown good optical and scintillation properties obtained thanks to the deep purification of ^{116}Cd and W, and the advantage of the low-thermal-gradient Czochralski technique to grow the crystal.

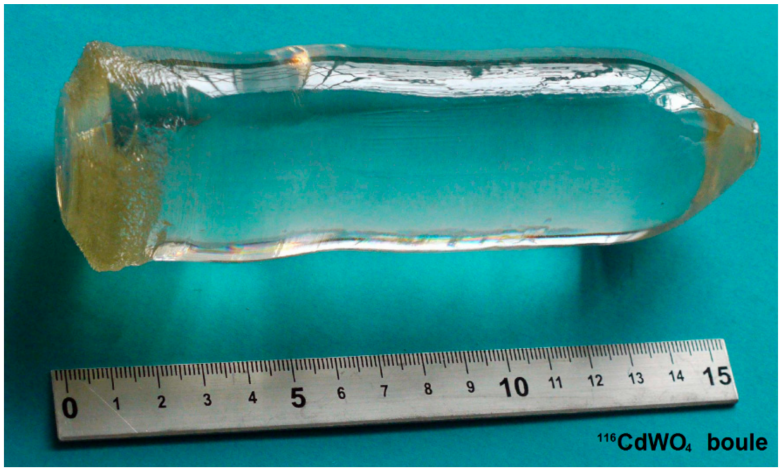

Figure 9. A large-mass crystal boule (~1.9 kg) of the enriched $^{116}CdWO_4$ crystal was used in the experiment Aurora [91].

The experiment with two cadmium tungsten crystal scintillators has been run in the low background DAMA/R&D set-up at the LNGS with several upgrades to improve the detector background counting rate and energy resolution [92–94]. The radioactive contamination by radionuclides from U/Th chains and K of the scintillating elements increases longitudinal during the crystal growth that shows a segregation of radio-impurities: The conic part of the boule, the beginning of the crystal growth, appeared the cleanest one. On Figure 10 the total energy spectrum accumulated with two $^{116}CdWO_4$ detectors over 26,831 h and selected by the pulse-shape and the front-edge analysis is shown: Raw data, spectra of γ(β), α, and ^{212}Bi-^{212}Po events (denoted "Bi-Po"). The spectra α(1) and α(2) demonstrate the distributions of alpha events accumulated by the detectors No. 1 from the upper part of the boule and No. 2, from the bottom part.

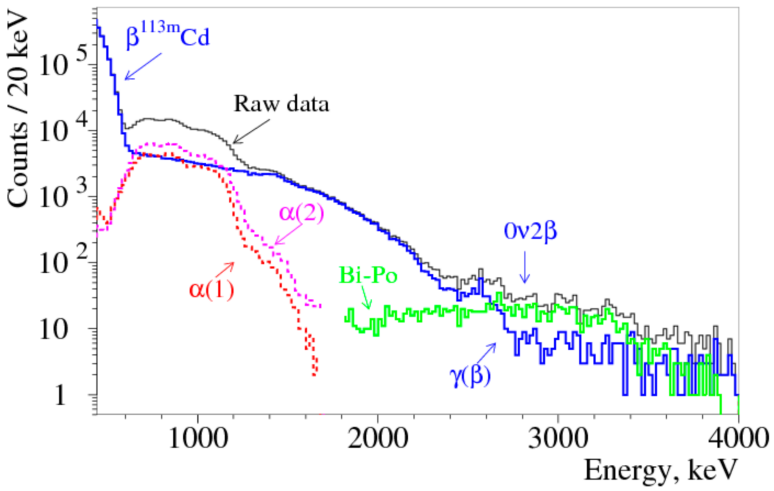

Figure 10. The sum energy spectrum accumulated with two $^{116}CdWO_4$ detectors over 26,831 h (Raw data) and spectra of γ(β), α and ^{212}Bi-^{212}Po events (denoted "Bi-Po") selected by the pulse-shape analysis. The spectra α(1) and α(2) denote the distributions of alpha events accumulated by the detectors No. 1 from the upper part of the boule and No. 2, from the bottom part.

The analysis of the recrystallized bottom part of the boule [64] shows that the application of an additional crystallization after the remelting of the grown crystal boule (double crystallization procedure) helps to reduce the radioactive contamination of the crystal. This feature is important for the production of radiopure crystal scintillators, especially from expensive enriched materials.

The results of the Aurora experiment to investigate 2β processes in ^{116}Cd with 1.162 kg of $^{116}CdWO_4$ scintillators, enriched in up to 82% in ^{116}Cd, are presented in [32]. In particular, the most accurate value of the half-life of ^{116}Cd relative to the 2ν2β decay to the ground state of ^{116}Sn was set: $T_{1/2}^{2\nu2\beta} = 2.63^{+0.11}_{-0.12} \times 10^{19}$ years, as well as a new half-life limit on the 0ν2β decay of ^{116}Cd to the ground state of ^{116}Sn: $T_{1/2}^{0\nu2\beta} > 2.2 \times 10^{23}$ year at 90% C.L. that corresponds to the limit on the effective Majorana neutrino mass $\langle m_\upsilon \rangle \leq (1.2-1.5)$ eV (depending on the nuclear matrix elements used in the estimations of the neutrino mass limit). Other 2β processes, including 0ν2β decay with different majorons (χ^0) emission were restricted at level 10^{20}–10^{22} years. The theoretical predictions for the transitions in ^{116}Cd are on the level of $T_{1/2} \sim 10^{21}-10^{24}$ years (see [95,96]). The best half-life limits on 2β processes in ^{116}Cd are presented in Table 3 [32].

Table 3. The best half-life limits on 2β processes in ^{116}Cd [32]. (Here, *n* defines the spectral index and *LV* denotes Lorentz-violating decay.)

Decay Channel, Level of ^{116}Sn (keV)	lim$T_{1/2}$(years) at 90% C.L.
2ν2β, g.s.	$2.63^{+0.11}_{-0.12} \times 10^{19}$
2ν2β, 2^+(1294)	$\geq 2.3 \times 10^{21}$ [97]
2ν2β, 0^+(1757)	$\geq 5.9 \times 10^{21}$ [97]
2ν2β, 0^+(2027)	$\geq 2.0 \times 10^{21}$ [97]
2ν2β, 2^+(2112)	$\geq 2.5 \times 10^{21}$
2ν2β, 2^+(2225)	$\geq 7.5 \times 10^{21}$
0ν2β, g.s.	$\geq 2.2 \times 10^{23}$
0ν2β, 2^+(1294)	$\geq 7.1 \times 10^{22}$
0ν2β, 0^+(1757)	$\geq 4.5 \times 10^{22}$
0ν2β, 0^+(2027)	$\geq 3.1 \times 10^{22}$
0ν2β, 2^+(2112)	$\geq 3.7 \times 10^{22}$
0ν2β, 2^+(2225)	$\geq 3.4 \times 10^{22}$
0νχ^0 $n = 1$, g.s.	$\geq 8.5 \times 10^{21}$ [98]
0νχ^0 $n = 2$, g.s.	$\geq 4.1 \times 10^{21}$
0νχ^0 $n = 3$, g.s.	$\geq 2.6 \times 10^{21}$
0ν$\chi^0\chi^0$ $n = 3$, g.s.	$\geq 2.6 \times 10^{21}$
2ν$\chi^0 LV$ $n = 1$, g.s.	$\geq 1.2 \times 10^{21}$
0ν$\chi^0\chi^0$ $n = 7$, g.s.	$\geq 8.9 \times 10^{20}$

$^{116}CdWO_4$ crystals can be used also as bolometers: High energy resolution and particle discrimination capability were tested at milli-Kelvin temperature with a 34.5 g enriched $^{116}CdWO_4$ sample with the sensitivity on the level of ~0.1 mBq/kg for ^{232}Th, ^{235}U, and ^{238}U (and their daughters) [99].

In addition, the same crystals that were used in the AURORA experiment, operated recently as scintillating bolometers in new measurements in two underground laboratories: One at the Laboratoire Souterrain de Modane, France (LSM) and another one at Laboratorio Subterraneo de Canfranc, Spain (LSC). Promising results were shown in regards to using them as bolometers with high energy resolution (11–16 keV at 2615 keV) and extremely high PSD for α and γ(β) particles (up to ~20σ) (see [56]).

6. Conclusions

Crystal scintillators, in particular, with a high concentration of the isotopes of interest, are powerful detectors for 2β experiments thanks to the low radiopurity level

and the possibility to implement calorimetric "source = detector" approach with a high detection efficiency. The deep purification of the initial materials, application of the low-thermal-gradient Czochralski technique, which provides a large volume of radiopure crystal scintillators with high optical quality and low losses, as well as the development of isotopically enriched crystals, are strongly required for the 2β decay investigations. The best half-life limit on the $2\nu\varepsilon\beta^+$ decay in ^{106}Cd to the ground state with the help of $^{106}CdWO_4$ crystal (216 g) enriched in ^{106}Cd up to 66.4% was set as $T_{1/2}^{2\nu\varepsilon\beta+} > 2.1 \times 10^{21}$ years, which reached the region of theoretical predictions. In the AURORA experiment, enriched in 82% in ^{116}Cd $^{116}CdWO_4$ scintillators, the most accurate value on the half-life of ^{116}Cd relative to the 2ν2β decay to the ground state of ^{116}Sn ($T_{1/2}^{2\nu2\beta} = 2.63^{+0.11}_{-0.12} \times 10^{19}$ years) and a new half-life limit on the 0ν2β decay of ^{116}Cd to the ground state of ^{116}Sn ($T_{1/2}^{0\nu2\beta} > 2.2 \times 10^{23}$ years at 90% C.L.) were set. The search for the 0ν mode of the 2β decay could be provided by using enriched crystal scintillators as bolometers that met the requirements of sensitive experimental equipment capable of registering extremely rare nuclear decays with half-lives of 10^{25}–10^{28} years (the possibility of using scintillation crystals enriched in ^{116}Cd as bolometers is under development).

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